Identifying the Sources of Subsurface Uranium Contamination at the Hanford Site, Washington

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RESEARCH OBJECTIVES

In the mid-1990s, a groundwater plume of uranium (U) and other contaminants was recognized in monitoring wells in the B-BX-BY Waste Management Area (WMA) at the Hanford Site in Washington. This area had been used since the late 1940s to store high-level radioactive waste and other products of U fuel-rod processing. Consequently, within a small area there are a number of potential sources for contamination. Some records exist of inadvertent waste spills, and several locations of vadose zone contamination had been identified. Two of these plumes were cored in an effort to understand the extent and nature of the contamination. However, the pattern of U concentration in these cores did not fully resolve the source issue. We undertook a study of the variation in U isotopic composition within the vadose zone and groundwater U plumes to better understand the source and history of contamination, as well as its transport in the subsurface.

APPROACH

The isotopic composition of natural U contrasts with the variable isotopic composition of U from processed fuel rods. This variation can be used as a tracer and fingerprint of contamination. We applied a new technique using MC-ICP-MS to provide high-precision isotopic measurements of small U samples, generating $^{234}U/^{238}U$, $^{235}U/^{238}U$, and $^{236}U/^{238}U$ ratios. Samples were provided from two cores (E33-45 and E33-46; see inset to Figure 1) through vadose zone plumes of U contamination near two single-shelled tanks in the B-BX-BY WMA. Uranium from pore waters in the sediment samples were separated and analyzed for isotopic composition. Aliquots of groundwater samples taken from wells in the area (see inset to Figure 1) were also analyzed. These samples cover the known extent of the groundwater U contamination plume. The results of the isotopic analyses are evaluated and compared to each other to identify sources and the extent of mixing with background U.

ACCOMPLISHMENTS

Twenty-five samples from the two cores were analyzed, along with nine groundwater samples. The results of the analyses are displayed in Figure 1. The isotopic signature of the E33-45 core contamination plume matches the most contaminated groundwater samples. In addition, the isotopic variation of the groundwater samples can be explained by mixing between natural background U and the E33-45 contamination U. The vadose zone plume in the E33-46 core was probably not a significant source of groundwater contamination.

SIGNIFICANCE OF FINDINGS

A comparison of the U isotopic data to model histories of fuel rod isotopic compositions demonstrates that the E33-45 contamination is consistent with a large spill event in 1951, as had been suspected from historical records. The locus of this spill is about 150 m from the core of the groundwater plume, suggesting significant horizontal displacement of the contamination before it encountered the water table. Over 50 years separates the vadose zone spill and the groundwater contamination detected in the mid-1990s, indicating a continuing large potential for contamination. Between 1993 and 2001, it appears that the groundwater U contamination migrated at an average of ~0.7 m/day. High-precision U isotopic measurements provide an improved tool for tracing and understanding the behavior of U contamination in the subsurface and groundwater.

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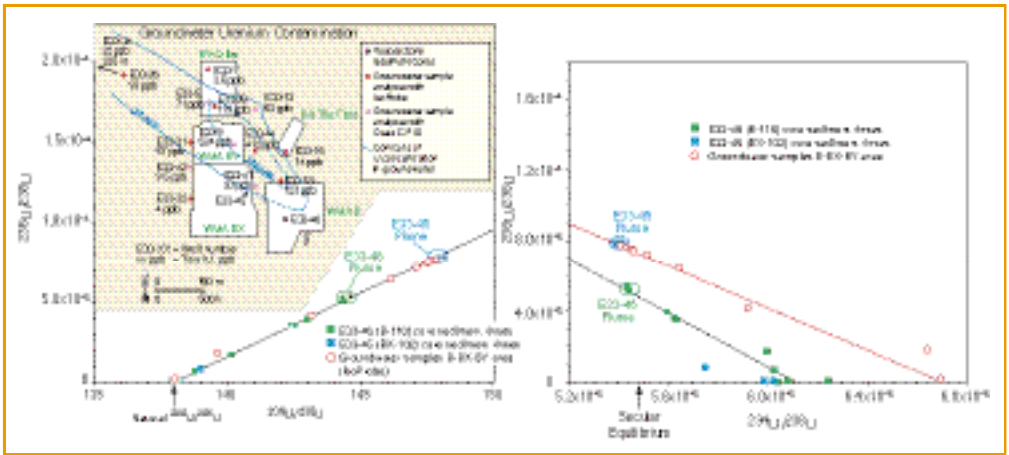

Figure 1. Uranium isotopic data for groundwater (red circles) samples, and sediment pore water from cores E33-45 (blue squares) and E33-46 (green squares). Left panel $^{238}U/^{235}U$ vs. $^{236}U/^{238}U$ and right panel $^{234}U/^{238}U$ vs. $^{236}U/^{238}U$. Inset in left panel is a map of analyzed sample well and core locations in the B-BX-BY Waste Management Area.